Japanese Journal of Applied Physics
Vol. 43, No. 2A, 2004, pp. L 180–L 182

Express Letter

Improved External Efficiency InGaN-Based Light-Emitting Diodes with Transparent Conductive Ga-Doped ZnO as p-Electrodes

Ken NAKAHARA*, Kentaro TAMURA, Mitsuhiko SAKAI, Daisuke NAKAGAWA,
Norikazu ITO, Masayuki SONOBE, Hidemi TAKASU, Hitoshi TAMPO[1], Paul FONS[1],
Koji MATSUBARA[1], Kakuya IWATA[1], Akimasa YAMADA[1] and Shigeru NIKI[1]

Optical Devices R&D Center, ROHM Co., Ltd., 21 Saiin Mizosaki-cho, Ukyo-ku, Kyoto 615-8585, Japan
[1]*Optoelectronic Materials and Devices Group, Photonics Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), Central 2, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan*

(Received November 18, 2003; accepted December 8, 2003; published January 9, 2004)

Transparent conductive Ga-doped ZnO (ZnO:Ga) was fabricated to serve as p-contacts of InGaN-based light-emitting diodes (LEDs) using molecular-beam epitaxy. As-grown ZnO:Ga films typically have resistivities of $\rho = 2 - 4 \times 10^{-4}\,\Omega\cdot\text{cm}$, and over 80% transparency in the near UV and visible wavelength ranges. The current-voltage characteristics between as-grown ZnO:Ga contacts and p-GaN layers were ohmic. The brightness of LEDs fabricated with ZnO:Ga p-contacts was nearly double compared to LEDs with conventional Ni/Au p-contacts. We obtained the external efficiency as high as 20.8% in the case of the near UV LED. The forward voltage at 20 mA was found not to increase even after the lamp LED with ZnO:Ga were kept for 80 h in high humidity and high temperature environments. [DOI: 10.1143/JJAP.43.L180]

KEYWORDS: ZnO, GaN, InGaN, LED, MBE, external efficiency, transparent, reliability, ohmic contact

Since the fabrication of high brightness and short wavelength light-emitting diodes (LEDs) based upon GaN, a large volume of research on GaN and its related materials have been performed. In particular, very bright white LEDs made using the combination of blue InGaN LEDs and $(Y_{1-a}Gd_a)_3(Al_{1-b}Ga_b)_5O_{12}:Ce^{3+}$ (YAG) phosphor[1] have opened the field of so-called solid-state lighting, and to date, many researchers have tried to substitute GaN-based LEDs for incandescent lamps or fluorescent light.

One of the most difficult obstacles in realizing solid-state lighting using GaN-based LEDs, however, is the lower luminescent efficiency of GaN-based LEDs as compared to fluorescent light tubes. The reasons for the low efficiency are low light extraction efficiency due to a small total reflection angle and the use of a broad p-electrode with low transparency. Broad p-contacts are indispensable to spread forward current over a wide area, because p-GaN is generally highly resistive and thus current spreading is not enough. The problem of low electrode transparency can be solved by using a highly transparent material for the p-electrodes. While there have been reports on application of indium tin oxide (ITO) as p-electrodes for InGaN-based LEDs, but some form of post-deposition annealing process was needed to obtain ohmic contact between the ITO and p-GaN layers and to obtain high transparency in many cases.[2–5]

Group-III-doped ZnO is another candidate for high transparent contact material. The *a*-axis lattice constant of ZnO is closely matched to that of GaN. ZnO can be grown epitaxially on GaN.[6–9] Thus low resistivity ZnO can serve as a more suitable electrode material than ITO for InGaN-based LEDs. However, there have been few reports on the application of ZnO as an n-[10] or a p-electrode material[11] for GaN based LEDs.

In this paper, we report that Ga-doped ZnO (ZnO:Ga) can be used as p-side ohmic contact material for InGaN LEDs without any post annealing processes. The use of ZnO:Ga improves the brightness of LEDs compared to those fabricated with conventional oxidized Ni/Au alloy electrodes. Furthermore, SiO_2 passivated InGaN LEDs with ZnO:Ga electrodes exhibit reliability at a level necessary for practical use.

InGaN-based LEDs were grown on *c*-plane sapphire substrates by metal-organic chemical vapor deposition. The precursors of Ga, In, and nitrogen were trimethyl indium, tirmethyl gallium, and ammonium, respectively. Si and Mg were used as dopants for n- and p-type films, respectively. A typical device structure is schematically shown in Fig. 1.

The sample preparation procedure before ZnO:Ga deposition was as follows. The GaN native oxide layers were removed in baths of 35% HCl solution for 1.5 min, samples were then rinsed with deionized water, and thermally cleaned at 400°C in a load lock vacuum chamber. ZnO:Ga films were deposited at a temperature of 300°C by a molecular-beam epitaxy (MBE) equipped with an oxygen radical source and Knudsen cells for Zn and Ga. Other machine parameters were the same as reported earlier.[12] The oxygen flow rate was 0.3 sccm with an RF power of 300 W.

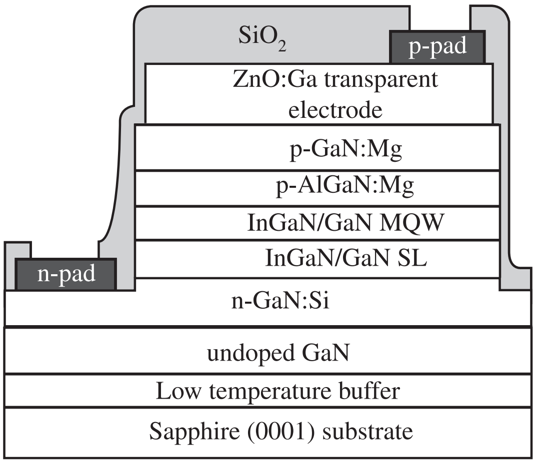

Fig. 1. Schematic cross-sectional structure of the InGaN-based light-emitting diode-chip with use of Ga-doped ZnO (ZnO:Ga) film as a p-contact electrode.

---

*E-mail address: optical_device@rohm.co.jp

The Zn flux supplied was fixed to be 2×10^{-6} Torr, and the Ga flux supplied varied up to 2×10^{-8} Torr. The Ga flux could not be measured when the Ga cell temperature was lower than 700°C, because the flux was lower than the background pressure ($\sim 1 \times 10^{-9}$ Torr).

After ZnO:Ga deposition, ZnO:Ga films were etched using diluted HCl solution, and the InGaN LED structure were partially etched to expose the n-GaN layer by use of reactive ion-beam etching. SiO_2 passivation layers were then deposited by plasma-chemical vapor deposition to cover whole LED chips after Ti/Al/Ti/Au pads were deposited on n-side and Ti/Au pads on the p-side by electron-beam evaporation for wire bonding. For comparison, we also prepared InGaN LEDs fabricated with conventional semi-transparent Ni (4 nm)/Au (8 nm) p-electrodes annealed at 600 °C for 5 min in N_2 and O_2 mixed ambient.

Hall measurements were carried out in the Van der Pauw configuration to evaluate the electrical properties of ZnO:Ga films deposited on double polished *a*-plane sapphire substrates. Table I shows the results of the Hall measurements. A resistivity of $\rho = 1.9 \times 10^{-4}$ Ω·cm was achieved. Figure 2 shows transmission spectra of the samples (c) and (d) of Table I and conventional oxidized Ni/Au alloyed metal. Over 80% transparency is retained in the wavelength range from 370 nm to 1000 nm, while the sample with Ni/Au contacts has a relatively lower transparency especially around 400 nm.

Table I. Properties of Ga-doped ZnO films grown on *a*-plane sapphire substrates. T_{Ga} and beam flux are a temperature of Knudsen cell of Ga and a Ga flux in a growth vacuum chamber, respectively. Electrical properties were characterized by Hall measurement with Van der Pauw configuration at room temperature. n_{Hall}, and ρ are carrier concentration and resistivity, respectively.

Specimen	T_{Ga} (°C)	Beam Flux (Torr)	n_{Hall} (cm^{-3})	ρ (Ω·cm)
a	650		2.9×10^{18}	1.1×10^{-1}
b	700		5.8×10^{19}	2.7×10^{-3}
c	750	4.0×10^{-9}	3.4×10^{20}	3.8×10^{-4}
d	800	1.8×10^{-8}	8.1×10^{20}	1.9×10^{-4}

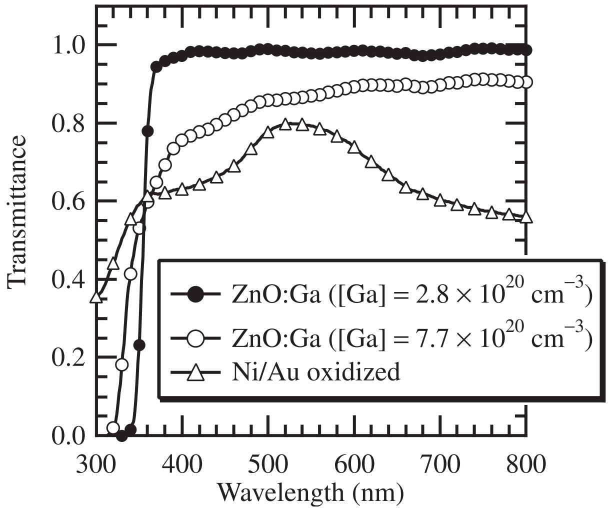

Fig. 2. Transmittance spectra of Ga-doped ZnO (ZnO:Ga) films grown on *a*-plane sapphire substrates and conventional Ni/Au oxidized alloy (open triangle) on a sapphire substrate. Ga concentrations ([Ga]) in ZnO, which were estimated by secondary ion mass spectroscopy, are 2.8×10^{20} cm^{-3} (closed circle) and 7.7×10^{20} cm^{-3} (open circle), respectively.

Figure 3 shows the current-voltage (*I-V*) characteristics between the as-deposited ZnO:Ga and p-GaN layers. The sample configuration is shown in the inset. As is clearly shown, the ZnO:Ga electrodes work as ohmic contacts for the p-GaN without any kind of post-deposition processing. The *I-V* characteristics of an InGaN LED are shown in Fig. 4. The forward voltage of an LED with ZnO:Ga electrodes is lower than that of an LED with conventional oxidized Ni/Au electrodes operated at the same current. This is an advantage in realizing high power efficiency and low heat generation, especially considering that over 100 mA forward current is necessary for solid-state lighting applications.

The current-brightness characteristics are shown in Fig. 5.

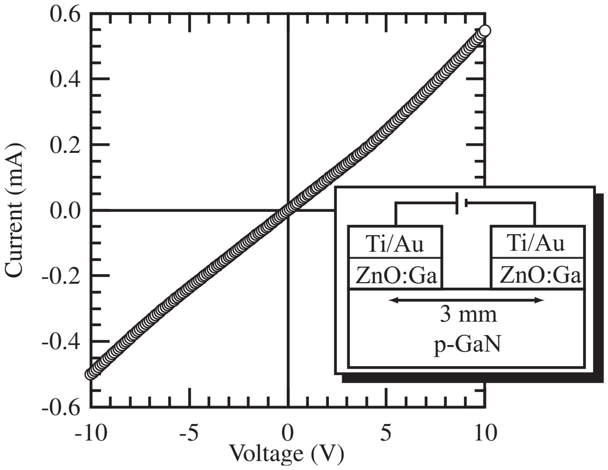

Fig. 3. Current-voltage characteristics between Ga-doped ZnO (ZnO:Ga) p-contacts and the p-GaN layer. Inset shows schematic configuration. Ti/Au pad electrodes ($\phi \sim 100\,\mu m$) were deposited on ZnO:Ga. Distance between contact electrodes is 3 mm.

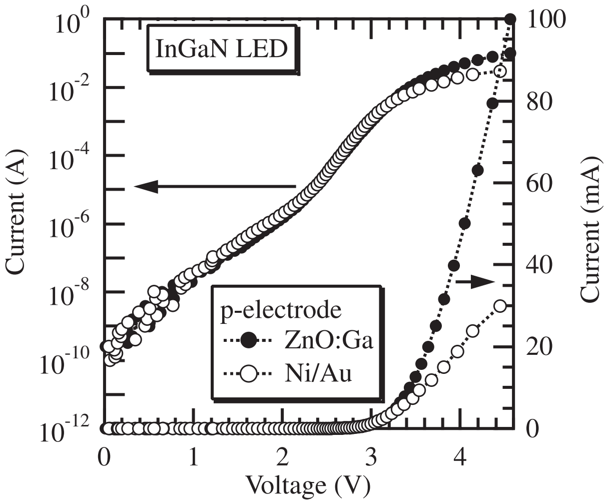

Fig. 4. Current-voltage characteristic of InGaN light-emitting diodes (LEDs) fabricated with Ga-doped ZnO (ZnO:Ga) p-contact (closed circle) and with Ni/Au-oxidized alloy p-contact (open circle), respectively. Left and right axes are logarithmic and liner scales, respectively.

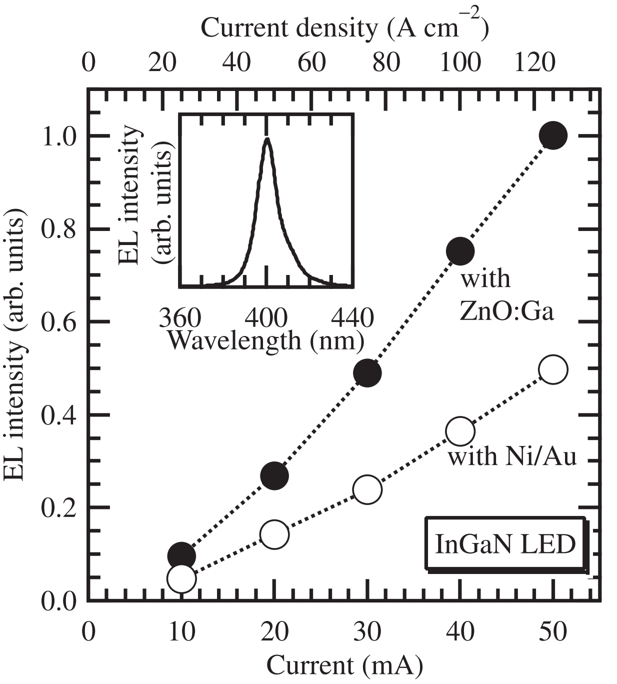

Fig. 5. Current (bottom axis) or current density (top axis) dependence of electro-luminescence (EL) intensity for InGaN light-emitting diodes (LEDs) fabricated with Ga-doped ZnO (ZnO:Ga) p-contact (closed circle) and with Ni/Au-oxidized alloy p-contact (open circle), respectively. Inset shows EL spectrum of an InGaN LED with ZnO:Ga p-contact operated at 20 mA.

The inset shows the emission spectrum. In this case, the brightness was measured by a photo-diode located perpendicular to the p-GaN surface. The brightness for LEDs with ZnO:Ga electrodes was found to be nearly double that of conventional semi-transparent oxidized Ni/Au electrodes, leading to high external efficiency. We obtained the external efficiency as high as 20.8% for an LED with ZnO:Ga electrode operated at 20 mA in the case that the emission peak wavelength was around 400 nm.

We confirmed that the initial properties were improved by use of ZnO:Ga electrodes as mentioned above. In the next step, we carried out a pressure cooker test (PCT) to check the reliability of ZnO:Ga electrodes. Shell type assembled LED lamps fabricated from epoxy resin were employed. We kept the lamp LEDs in an environment in which the temperature was 121°C, the humidity 85%, and the pressure was 1.6 atm for 80 h, and measured the forward *I-V* characteristics before and after the test to evaluate device reliability. As clearly shown in Fig. 6, no significant changes were observed after the test, and therefore the reliability of ZnO:Ga p-electrodes is sufficient for practical use.

We deposited ZnO:Ga as p-electrodes for InGaN-based LEDs by use of MBE. The results are summarized as below.

(1) ZnO:Ga worked as ohmic contacts for p-GaN without any kind of post-deposition treatment.
(2) InGaN LEDs with ZnO:Ga electrodes had the lowest forward voltage of 3.5 V at 20 mA. This value is lower than that of LEDs with conventional oxidized Ni/Au p-contacts.

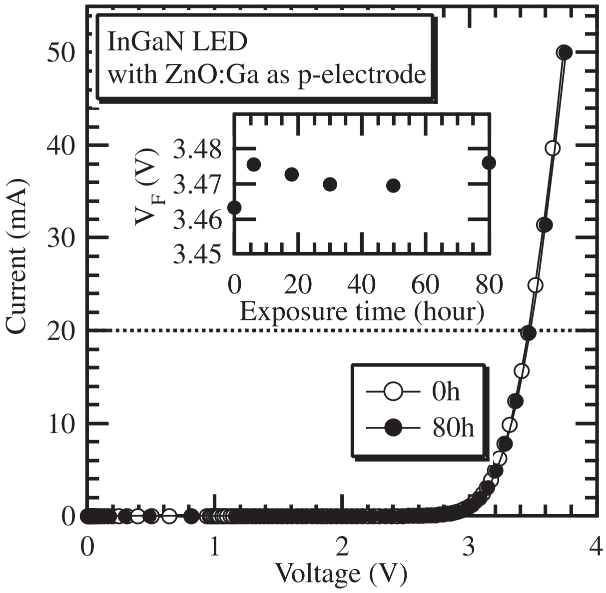

Fig. 6. Current-voltage (*I-V*) characteristics of an InGaN light-emitting diode (LED) lamp fabricated with Ga-doped ZnO (ZnO:Ga) p-contact. Reliability test, which called a pressure cooker test (PCT), was carried out in an environment in which the temperature was 121°C, the humidity 85%, and the pressure was 1.6 atm for 80 h (closed circle). An initial *I-V* profile (0 h) is also shown, for comparison (open circle). Inset shows the reliability-test exposure time dependence of forward voltage (V_F) of the lamp LED operated at 20 mA.

(3) The brightness increased nearly by a factor of 2 compared to LEDs with conventional oxidized Ni/Au p-electrodes.
(4) No significant change in forward I-V characteristics were observed after the PCT test.

1) S. Nakamura and G. Fasol: *The Blue Laser Diode: GaN Based Light Emitters and Lasers* (Springer, Berlin, 1997).
2) T. Margalith, O. Buchinsky, D. A. Cohen, A. C. Abare, M. Hansen, S. P. DenBaars and L. A. Coldren: Appl. Phys. Lett. **74** (1999) 3930.
3) D. W. Kim, Y. J. Sung, J. W. Park and G. Y. Yeom: Thin Solid Films **398-399** (2001) 87.
4) R. H. Horng, D. S. Wuu, Y. C. Lien and W. H. Lan: Appl. Phys. Lett. **79** (2001) 2925.
5) S. Y. Kim, H. W. Jang and J. L. Lee: Appl. Phys. Lett. **82** (2003) 61.
6) M. A. L. Johnson, S. Fujita, W. H. Rowland, Jr., W. C. Hughes, J. W. Cook, Jr. and J. F. Schetzina: J. Electron. Mater. **25** (1996) 855.
7) R. D. Vispute, V. Talyansky, S. Choopun, R. P. Sharma, T. Venkatesan, M. He, X. Tang, J. B. Halpern, M. G. Spencer, Y. X. Li, L. G. Salamanca-Riba, A. A. Iliadis and K. A. Jones: Appl. Phys. Lett. **73** (1998) 348.
8) S. K. Hong, H. J. Ko, Y. F. Chen and T. Yao: J. Cryst. Growth **209** (2000) 537.
9) H. J. Ko, Y. F. Chen, S. K. Hong, H. Wenisch, T. Yao and D. C. Look: Appl. Phys. Lett. **77** (2000) 3761.
10) C. T. Lee, Q. X. Yu, B. T. Tang, H. Y. Lee and F. T. Hwang: Appl. Phys. Lett. **78** (2001) 3412.
11) J. O. Song, K. K. Kim, S. J. Park and T. Y. Seong: Appl. Phys. Lett. **83** (2003) 479.
12) K. Nakahara, T. Tanabe, H. Takasu, P. Fons, K. Iwata, A. Yamada, K. Matsubara, R. Hunger and S. Niki: Jpn. J. Appl. Phys. **40** (2001) 250.